Sensitivity of sulfate direct climate forcing to the particle physical state: A global perspective

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Sulfate physical state and chemical composition

Neutralization $X= [NH_4]/2[SO_4]$

Solids:

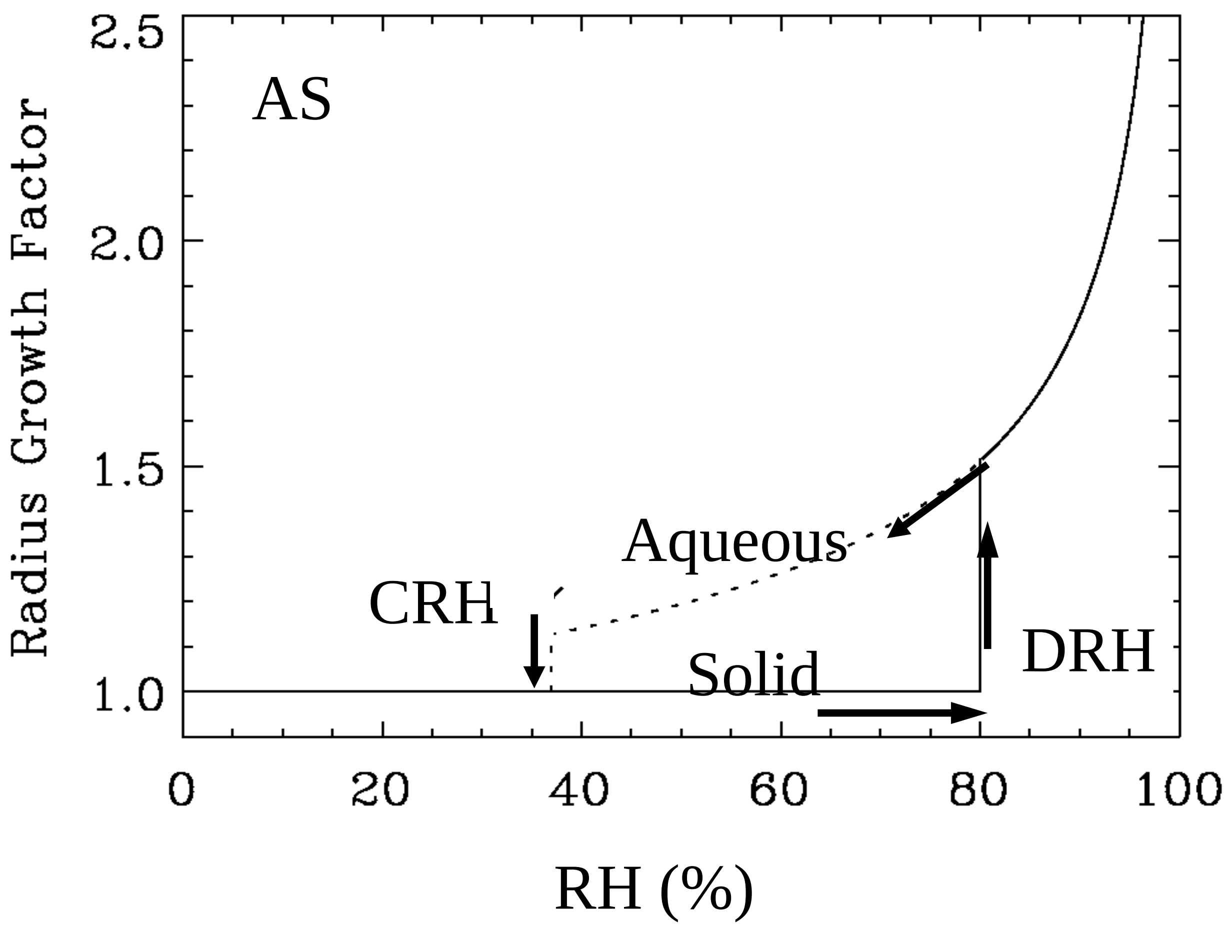
AS	$(NH_4)_2SO_4$	$X = 1$
LET	$(NH_4)_3H(SO_4)_2$	$X = 0.75$
AHS	$(NH_4)\ HSO_4$	$X = 0.5$

Aqueous:

SA	H_2SO_4, H_2O	$X = 0$
SO4aq	SO_4^{2-}, H^+, NH_4^+, H_2O	$0<X<1$

Hysteresis Loop of Sulfate Hygroscopicity

CRH: Crystalline relative humidity
DRH: Deliquesce relative humidity



3 factors:

(1) local RH
(2) RH history
(3) CRH & DRH

CRH<RH<DRH
solids or aqueous?

Composition-dependence of CRH and DRH

DRH:	always aqueous	40%	69%	80%
	SA	AHS	LET	AS
X:	0.0	0.5	0.75	1.0
CRH(X):	0	5%	30%	37%

Polynomials of X

Martin et al., 2003

An example:

aqueous particles with X = 0.9 (CRH(x)=32%) at RH = 85%

1) Decreasing RH to 70%, all aqueous
2) Decreasing RH to 32%, all solid particles (LET&AS).
3) Increasing RH to 70%, mixed phase (aqueous LET and solid AS)

Importance of Sulfate Physical State

- Microphysical importance
  - Particular Matter (PM) Air quality (particle size and mass)
  - Heterogeneous chemistry (hydrolysis of N_2O_5)
  - Cloud formation (ice/water CCN)

- Radiative importance
  - Aerosol refractive index and size
  - Sulfate direct climate forcing (SDCF)
  - Visibility

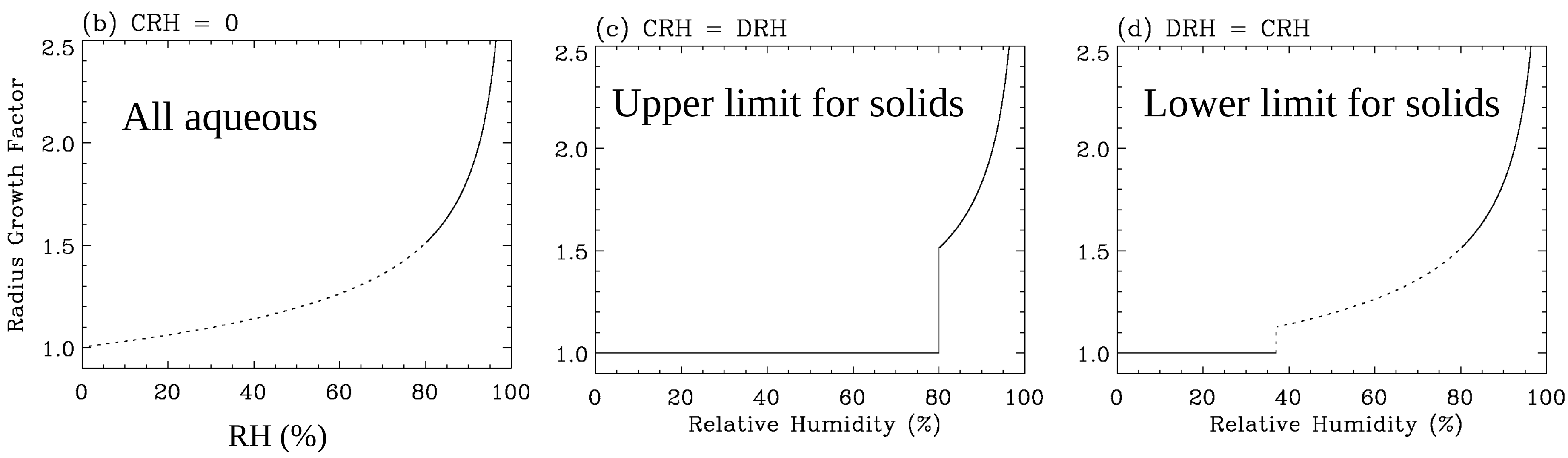
Previous Studies on Sulfate Phase Transition

CTM:

1) No phase, only SO_4 mixing ratio
2) Diagnosis phase based on local X and RH, with assumed RH history.
3) track RH history using Lagrangian model, and diagnosis the phase.

Radiative calculation:

1) CRH and DRH equal to a RH threshold to remove bifurcation.
2) Other parameterization methods



An example

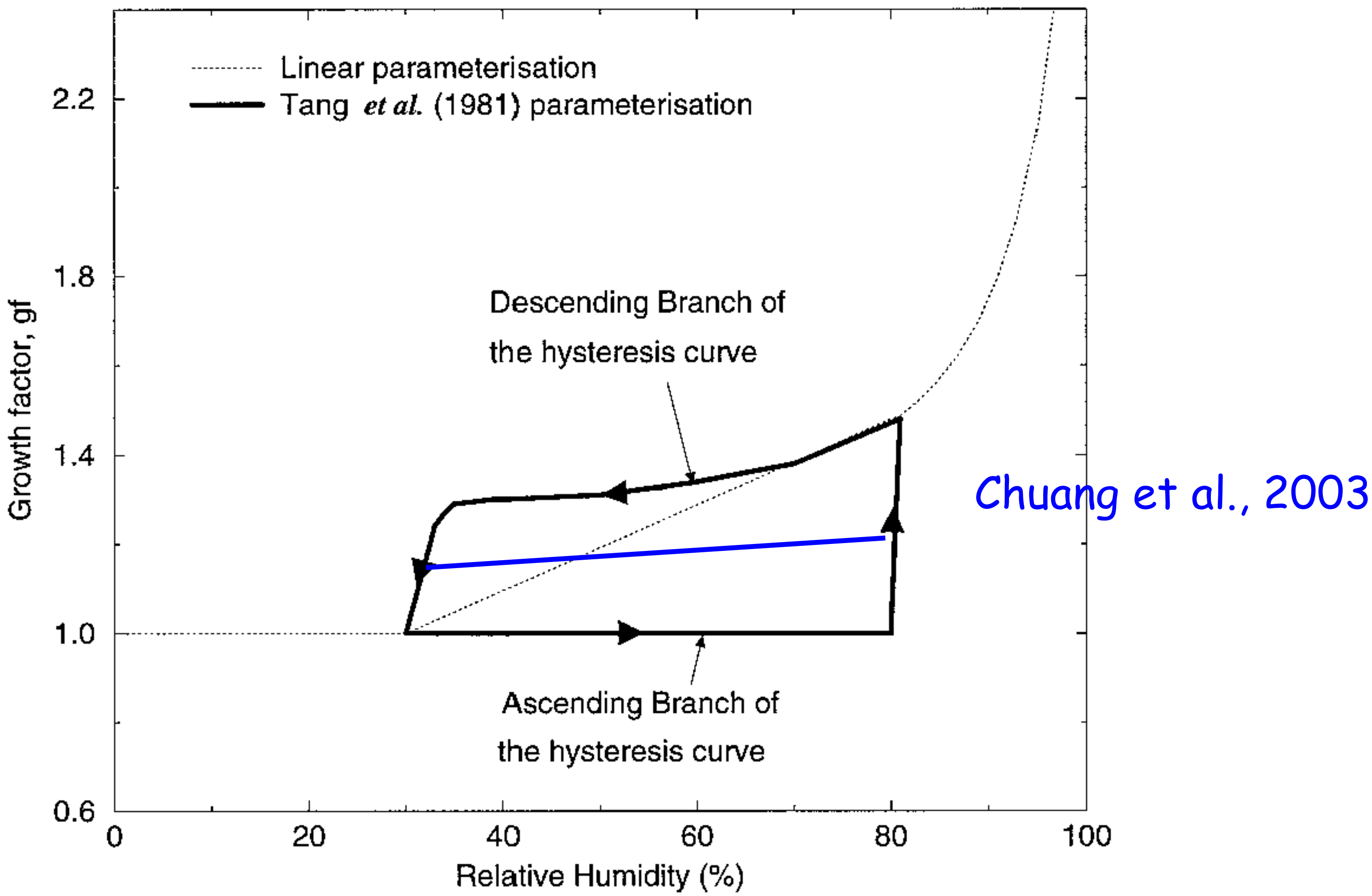


FIG. 1. The growth factor for ammonium sulfate aerosol as a function of relative humidity for the linear, ascending, and descending relative humidity schemes described in the text.

Haywood et al., 1997
with modification

Uncertainty Envelope from Previous Studies

Assuming CRH and DRH of AS for all sulfate particles

Aerosol type	Levels	Relative humidity parameterization	Cloud scheme	NH W m^{-2}	SH W m^{-2}	Global W m^{-2}	Global change %
$(NH_4)_2SO_4$	7	linear	UM	−0.60	−0.15	−0.38	0
$(NH_4)_2SO_4$	7	linear	cloud mask	−0.59	−0.14	−0.36	−5
$(NH_4)_2SO_4$	5	linear	UM	−0.67	−0.17	−0.42	+11
$(NH_4)_2SO_4$	10	linear	UM	−0.57	−0.14	−0.36	−5
$(NH_4)_2SO_4$	7	dry	UM	−0.38	−0.09	−0.24	−37
$(NH_4)_2SO_4$	7	descending	UM	−0.63	−0.16	−0.39	+3
$(NH_4)_2SO_4$	7	ascending	UM	−0.54	−0.14	−0.34	−11
H_2SO_4	7	d'Almeida et al. (1991)	UM	−0.66	−0.17	−0.41	+8

Haywood et al., 1997

Using CRH and DRH from aerosol thermodynamical model.

(F_L and F_U, respectively, in W m^{-2}) are calculated. Including both anthropogenic and natural emissions, we obtain global annual averages of $F_L = -0.750$, $F_U = -0.930$, and

$\Delta F_{U,L}$=24% for full sky calculations without clouds and $F_L = -0.485$, $F_U = -0.605$, and $\Delta F_{U,L}$=25% when clouds are included. Regionally, $\Delta F_{U,L}$=48% over the USA, 55%

Martin et al, 2004.

Questions

1) Mass percentage of solid sulfate?
2) Contribution of solids to sulfate direct climate forcing (DCF) ?

A systematic bias or error (not ± random uncertainty) !

Approach

1) A box model approach
2) GEOS-CHEM investigation

Box Model Estimate

Under thin-layer approximation (*Wiscombe and Grams*, 1976):

$$SDCF = -A\,(\omega_{sd}\ \overline{\beta}_{sd}\ \tau_{sd} + \omega_{aq}\ \overline{\beta}_{aq}\ \tau_{aq})$$

Solid Aqueous

$$A = -\frac{1}{2} S_0 T(1 - A_c)(1 - R_s)^2$$

S_0: solar constant

A_c: cloud fraction

R_s: surface reflectance

T: atmos. transmittance

ω: single scattering abledo (= 1)

$\overline{\beta}$: time-averaged backscattering fraction

τ: aerosol optical thickness

Climate System parameters

Forcing Agent parameters

Importance of Solids on Forcing Estimate

Aerosol term: $\overline{\beta}_{sd}\ \tau_{sd} + \overline{\beta}_{aq}\ \tau_{aq}$

$$= \overline{\beta}_{sd} B_{sd} E_{sd} + \overline{\beta}_{aq} B_{aq} E_{aq}$$

Where
E: mass extinction efficiency $m^2\ (gSO4^{2-})^{-1}$
B: sulfate burden $(gSO4^{2-})\ m^{-2}$

B_{sd}/B_{aq} is unknown, previous studies use:

$$= \overline{\beta}'\ E'\ B$$

where $\overline{\beta}'\ E' = \dfrac{\overline{\beta}_{sd}\ E_{sd} B_{sd} + \overline{\beta}_{aq}\ E_{aq} B_{aq}}{B}$

$$= \overline{\beta}'\ E_{sd}\ G_{\tau}\ B$$

$B = B_{sd} + B_{aq}$

Where

$$G_{\tau} = G_E - \frac{B_{sd}}{B}(G_E - 1)$$

$$G_E = \frac{E_{aq}}{E_{sd}}$$

Previous common assumption:

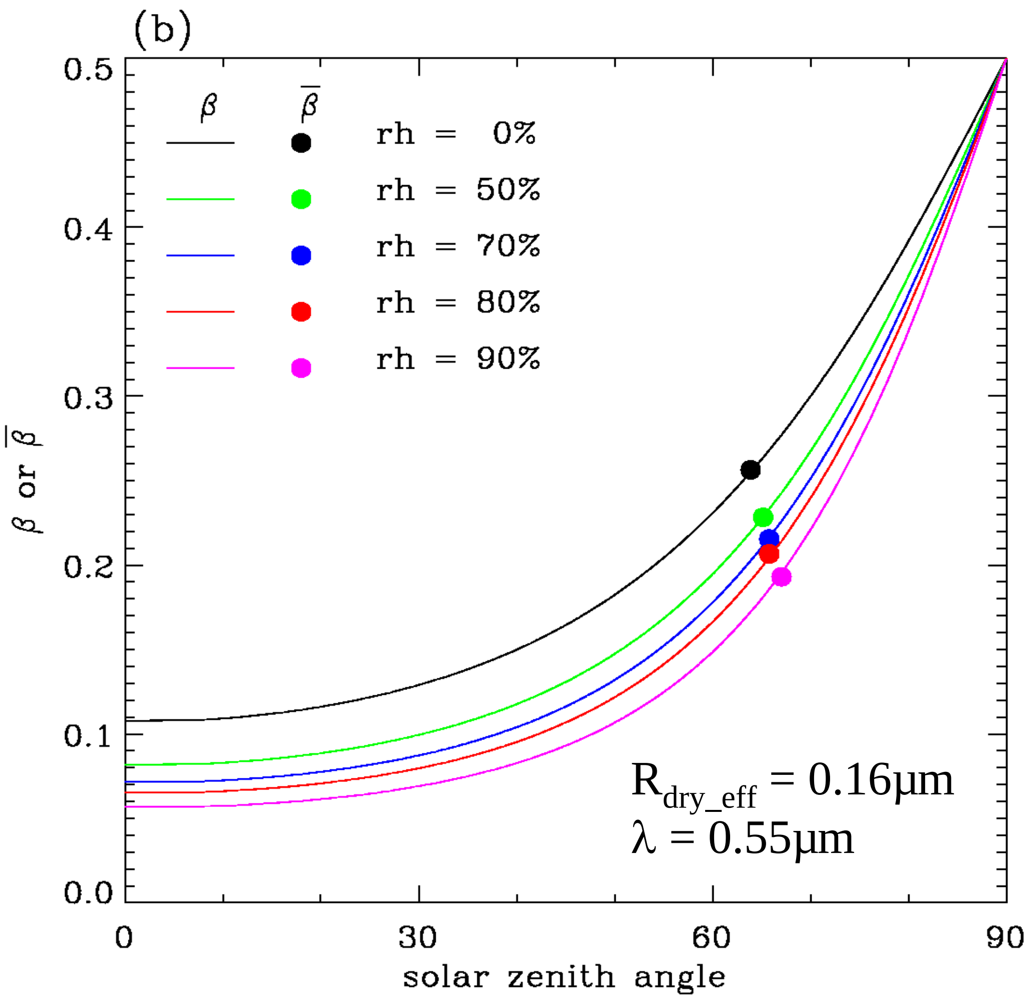
$G_{\tau} = G_E$

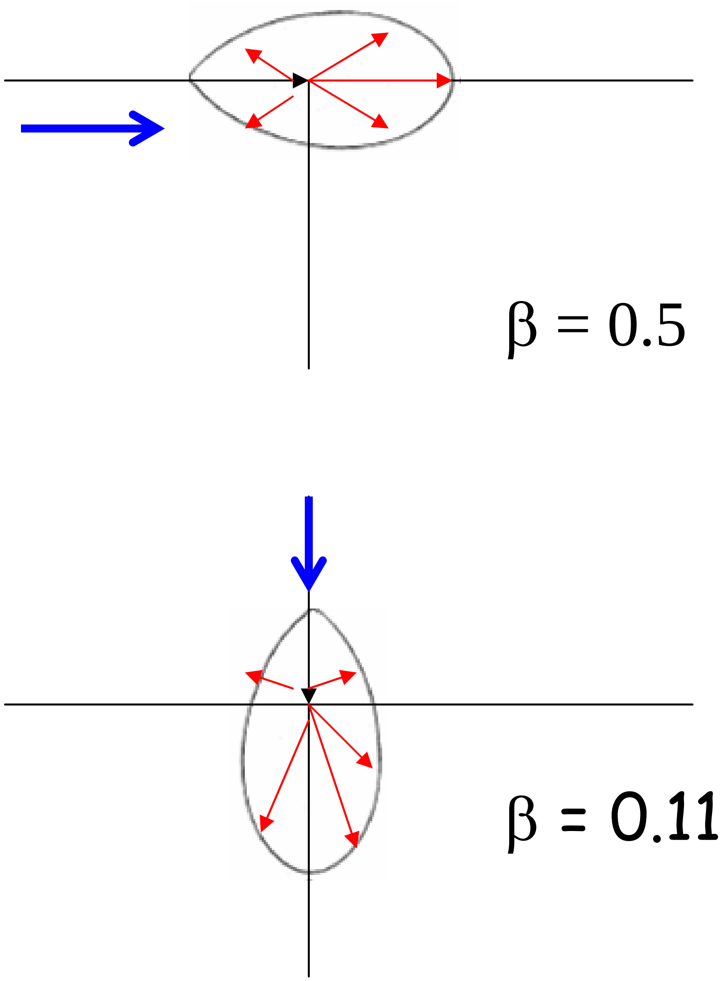
$\overline{\beta}' = \overline{\beta}_{sd}$ or $\overline{\beta}' = \overline{\beta}_{aq}$

G_{τ} : Optical thickness growth factor

$G_{E:}$ Mass extinction efficiency growth factor

$\overline{\beta}$: time-averaged backscattering fraction

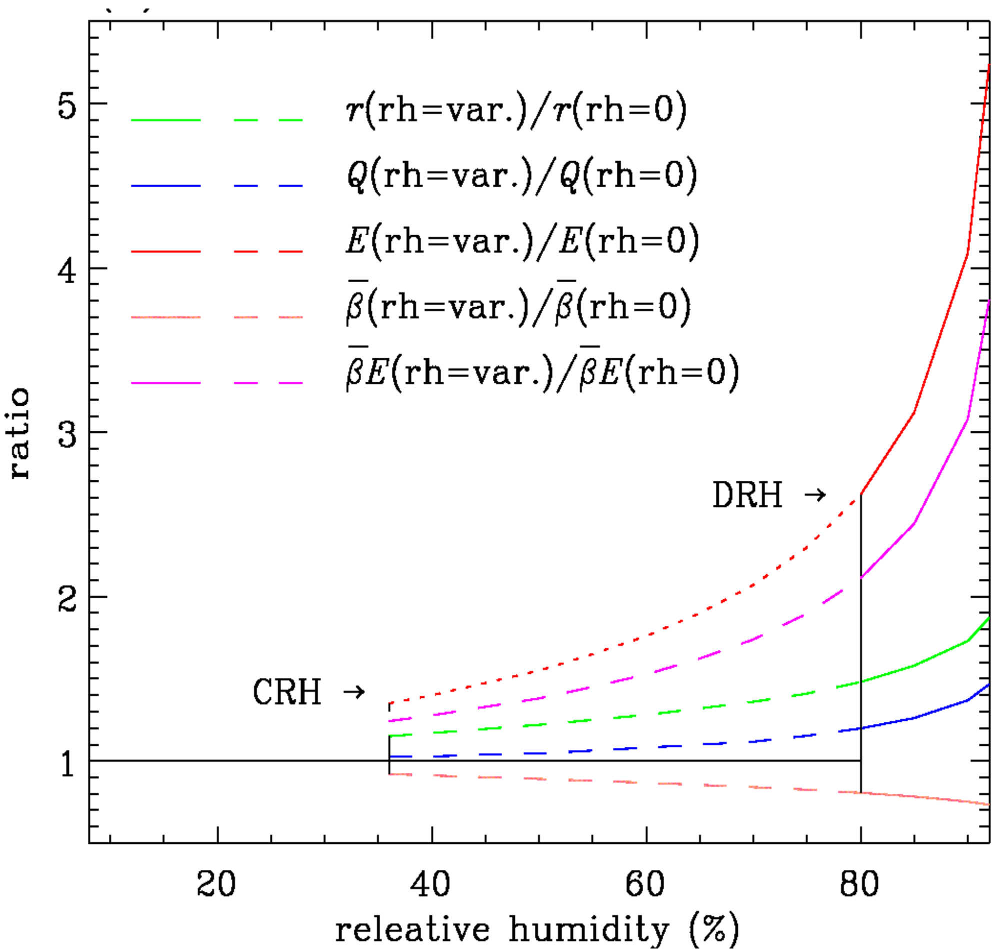




At RH = 80%, $\overline{\beta}_{aq} = 0.7\,\overline{\beta}_{sd}$

$$\overline{\beta} = \int_{\mu_{min}}^{\mu_{max}} \beta(\mu)\, d\mu$$

sulfate physical state impact on scattering properties



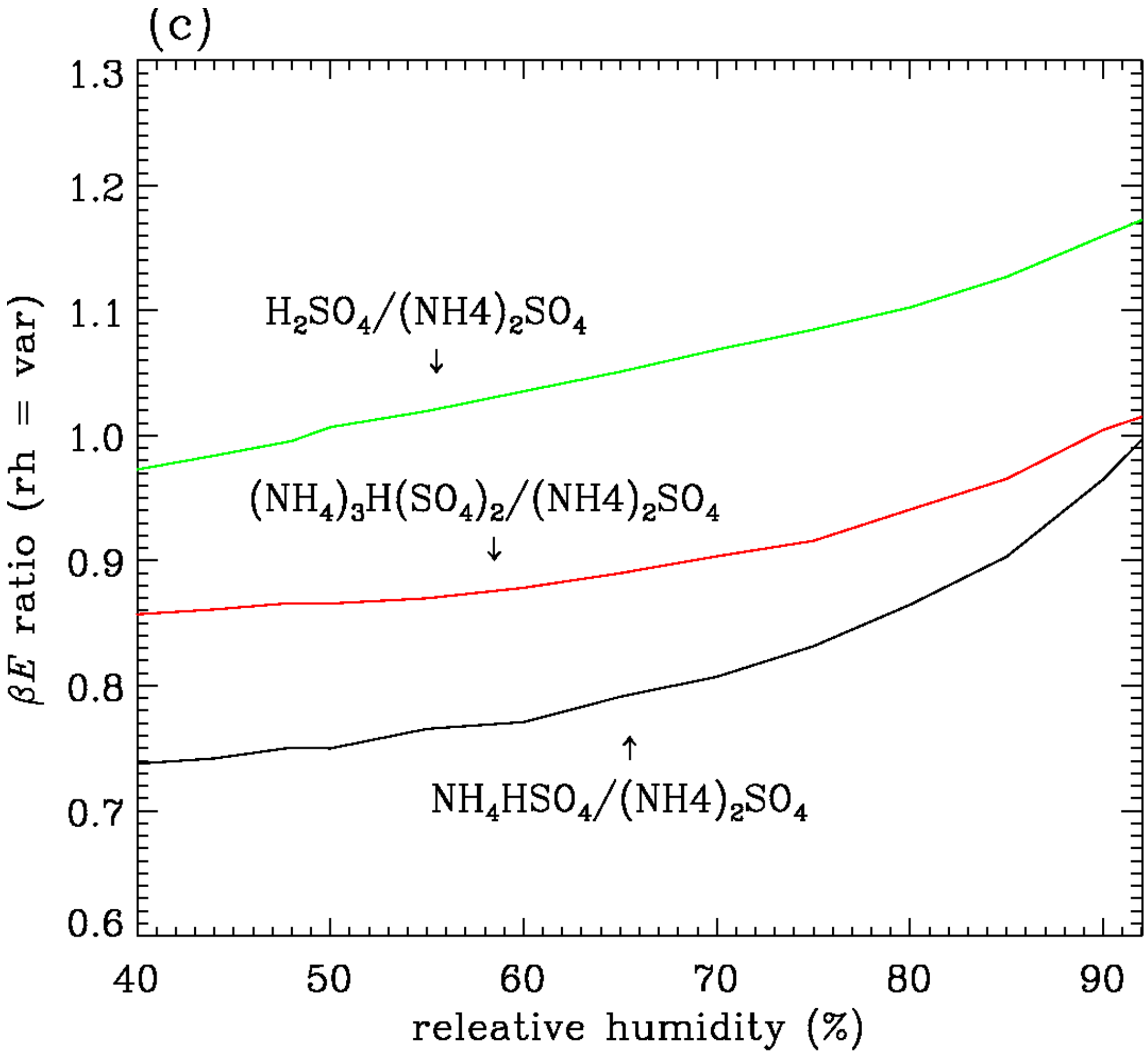
For aqueous particles, as RH increases,

r increases,
Q (extinction efficiency) and *E* increase

However, β decreases

Overall, increase the aerosol forcing.

Impact of Chemical Composition



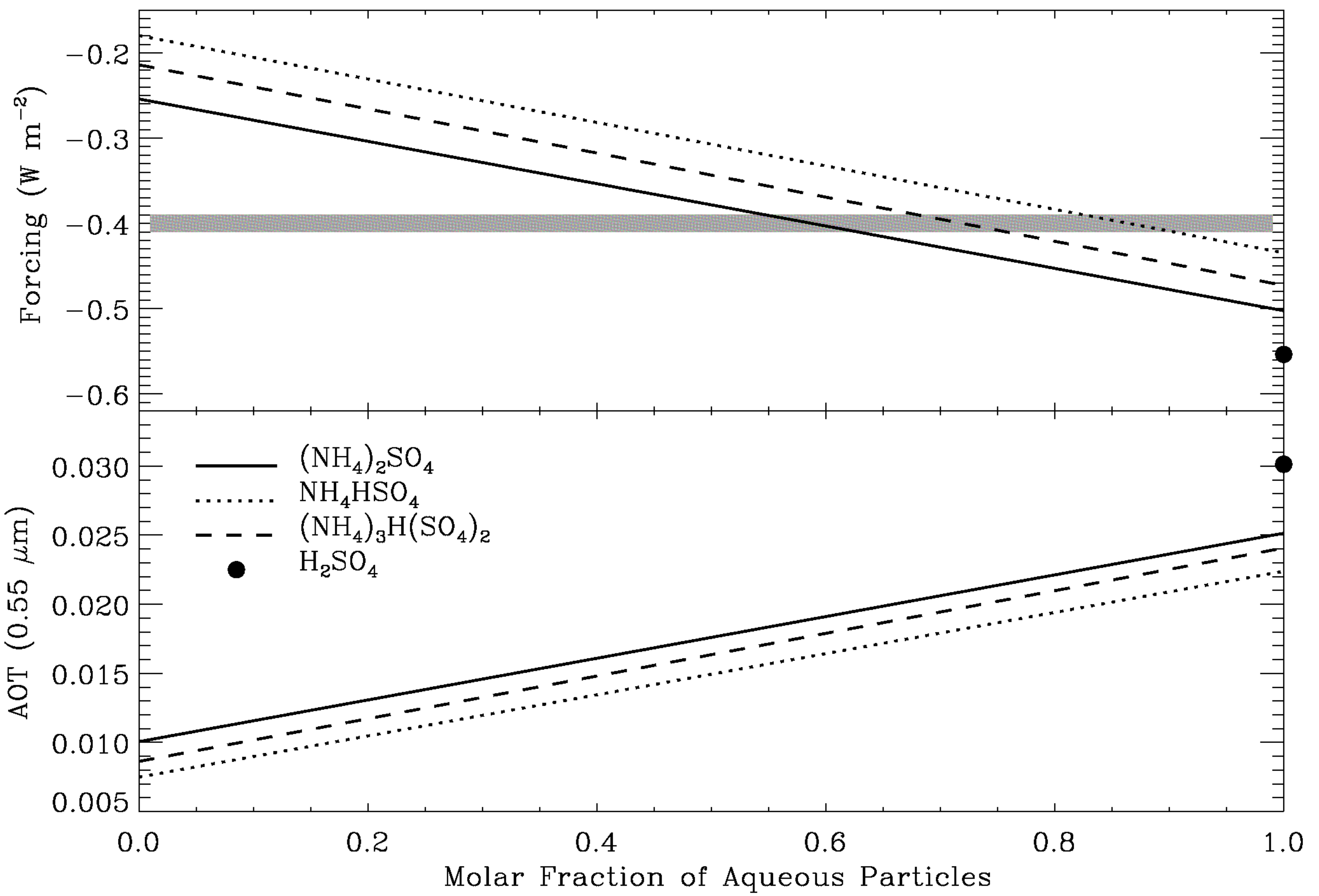
$\overline{\beta}_{aq}$ τ_{aq}

20% difference due to Δcomposition

$\overline{\beta}_{sd}$ τ_{sd}

30% difference due to Δcomposition

Sulfate AOT and forcing for variable B_{sd}/B



IPCC best estimate

The optical properties for aqueous particles at RH=80% is used. Climate system parameters are same as Charlson et al (1992)

GEOS-CHEM Investigation

1) Seasonal and geographical distribution of B_{sd}

2) Percentage of solid particle contribution to the global full-sky anthropogenic forcing

3) Forcing difference in the following 4 cases, basecase, CRH = 0, CRH=DRH, and DRH=CRH.

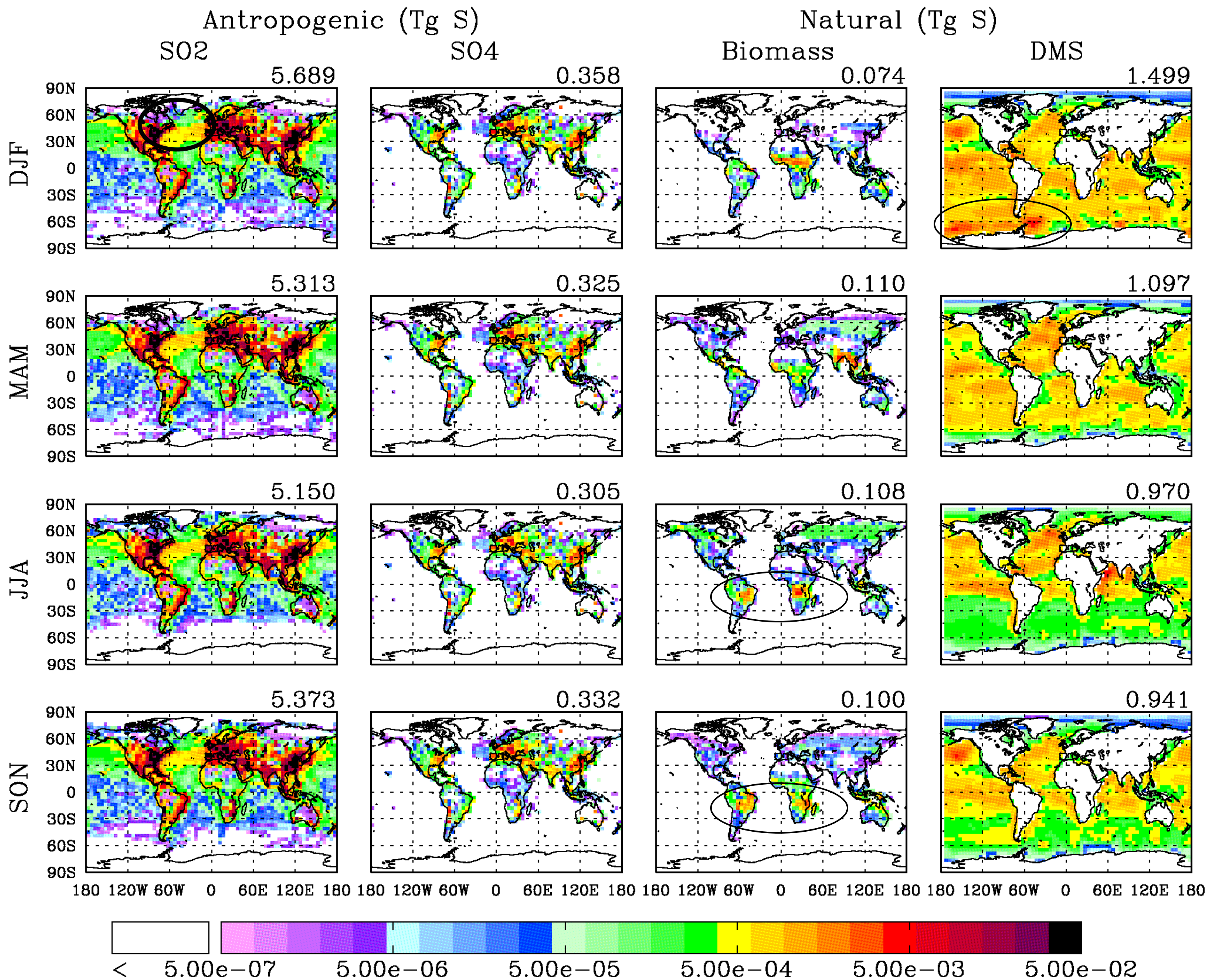
4) Where and how much would be largest forcing difference caused by the sulfate phase transition (regional perspective).

Model development

- Based upon Park et al (2004), with the following modification:
  - No nitrate. H_2SO_4-H_2O system only.
  - Sulfate mass are transported in 4 species: solid AS, LET, and AHS particles, aqueous SO_4.
  - CRH (X) and DRH of solids from Martin et al., 2003.
  - The concentration of each sulfate species are calculated according to the CRH(X), X, ambient RH, and DRH values.

- Because we explicitly track the solid and aqueous phase at each time step and model grid, the RH history on sulfate phase is retained.

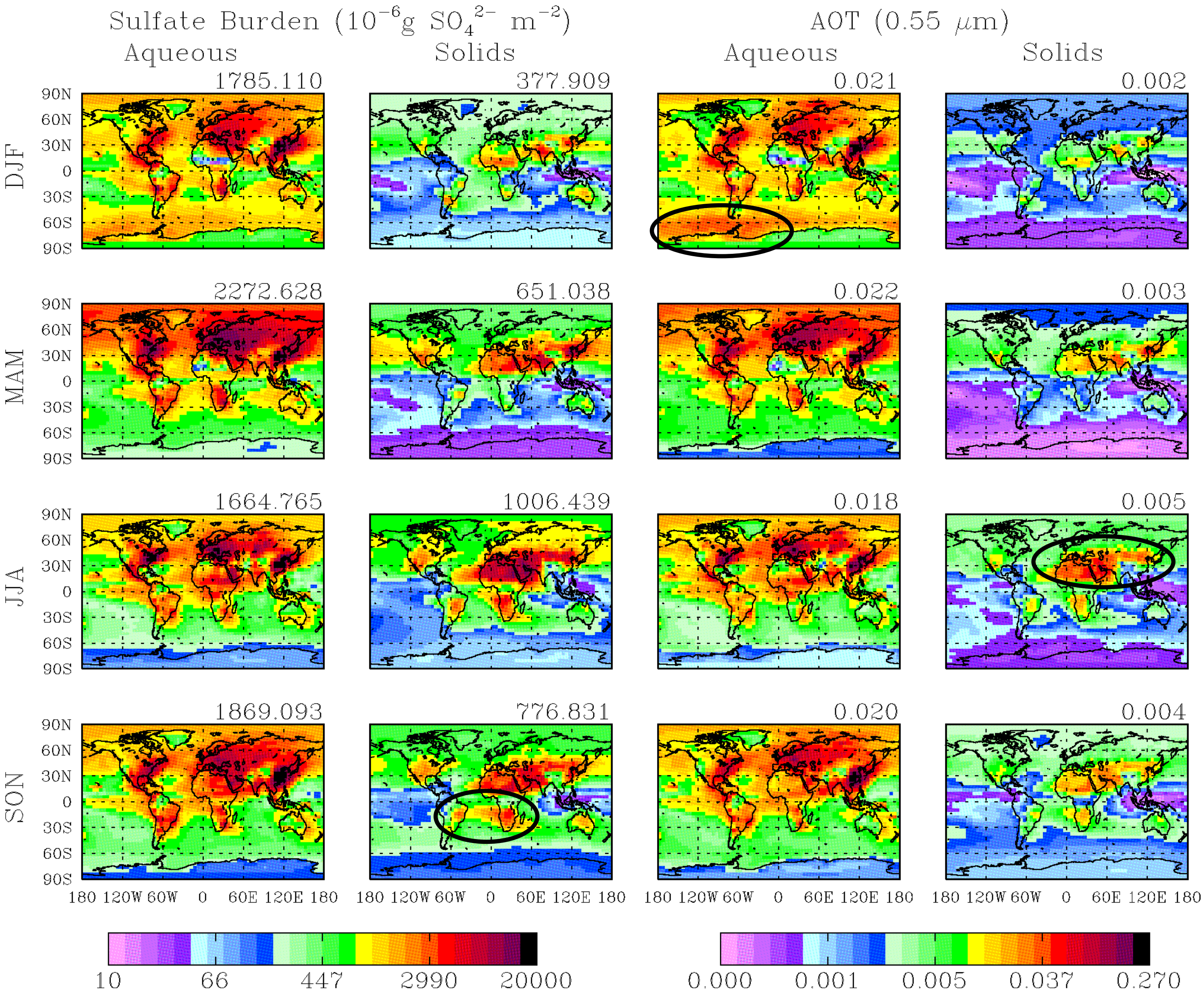
Emissions: ~80% are anthropogenic



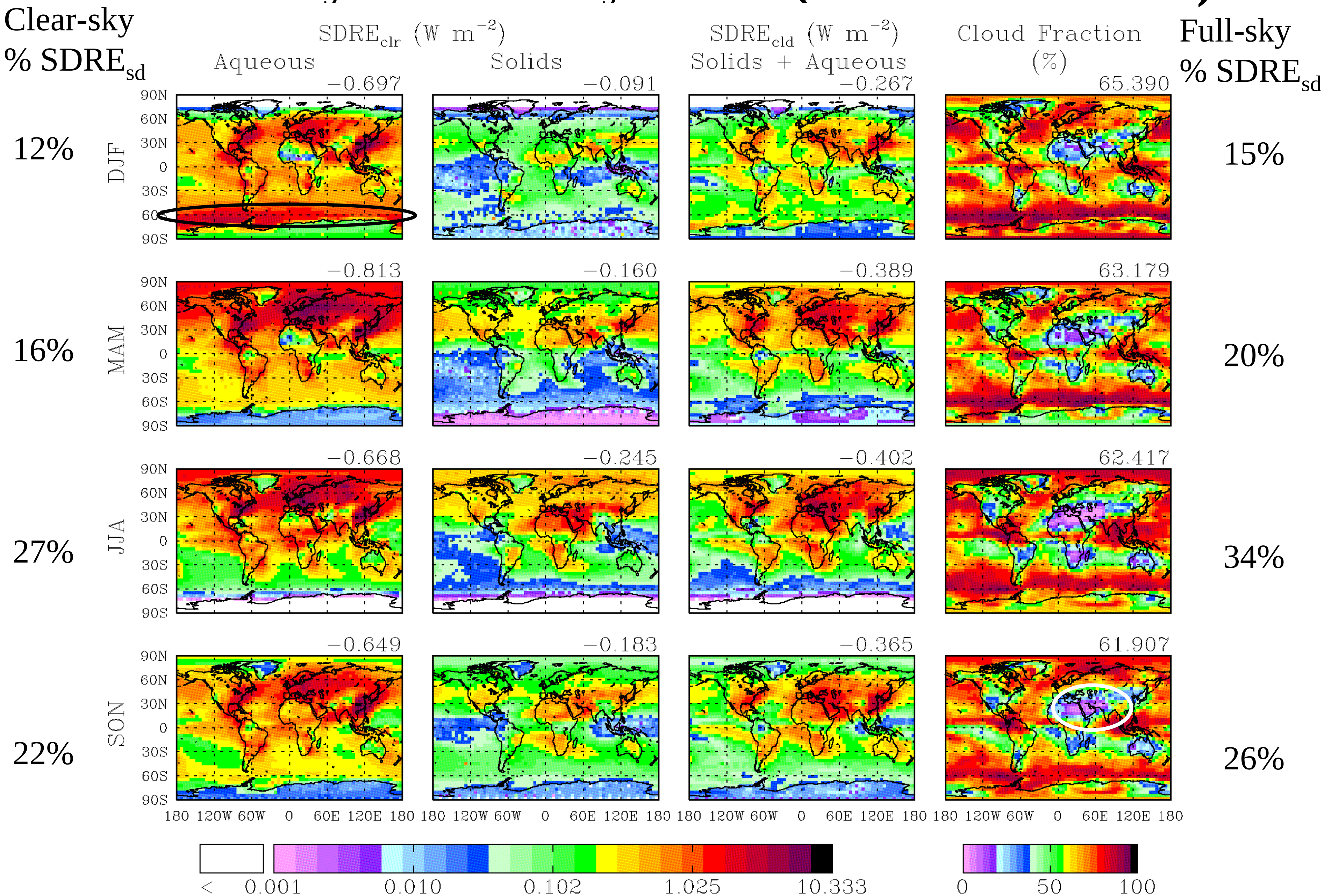
Following Park et al (2004), we consider biomass burning as natural emission.

Seasonal SO_4^{2-} burden and AOT in base case

% B_{sd} | % τ_{sd}
17% | 9%
22% | 12%
38% | 22%
29% | 17%



Clear-sky and full-sky SDRE (natural + anthro.)



Covariance matters. Cloud distribution favors the forcing of solids.

SDRF (~70% of SDRE)

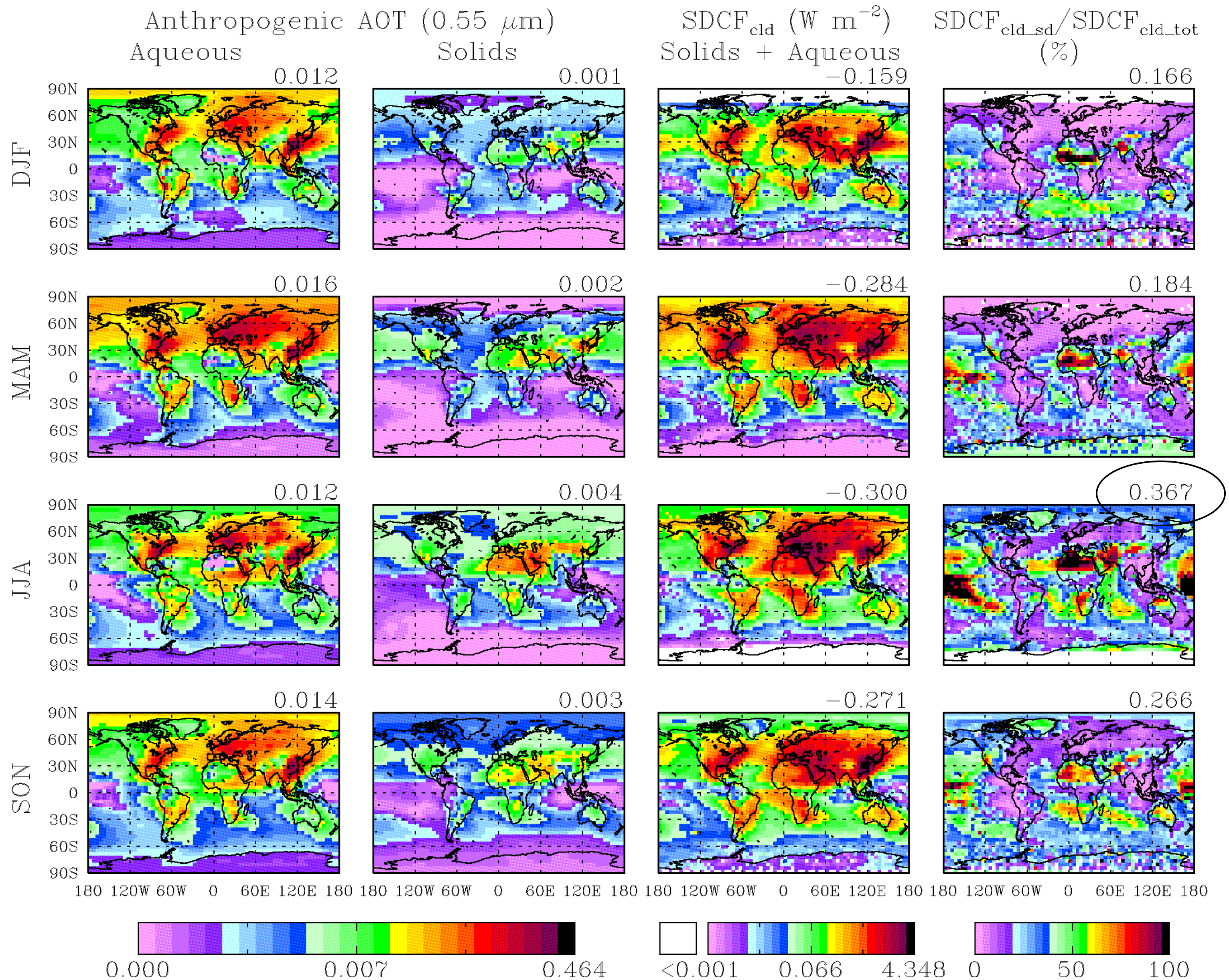
% τ_{sd}

8%

11%

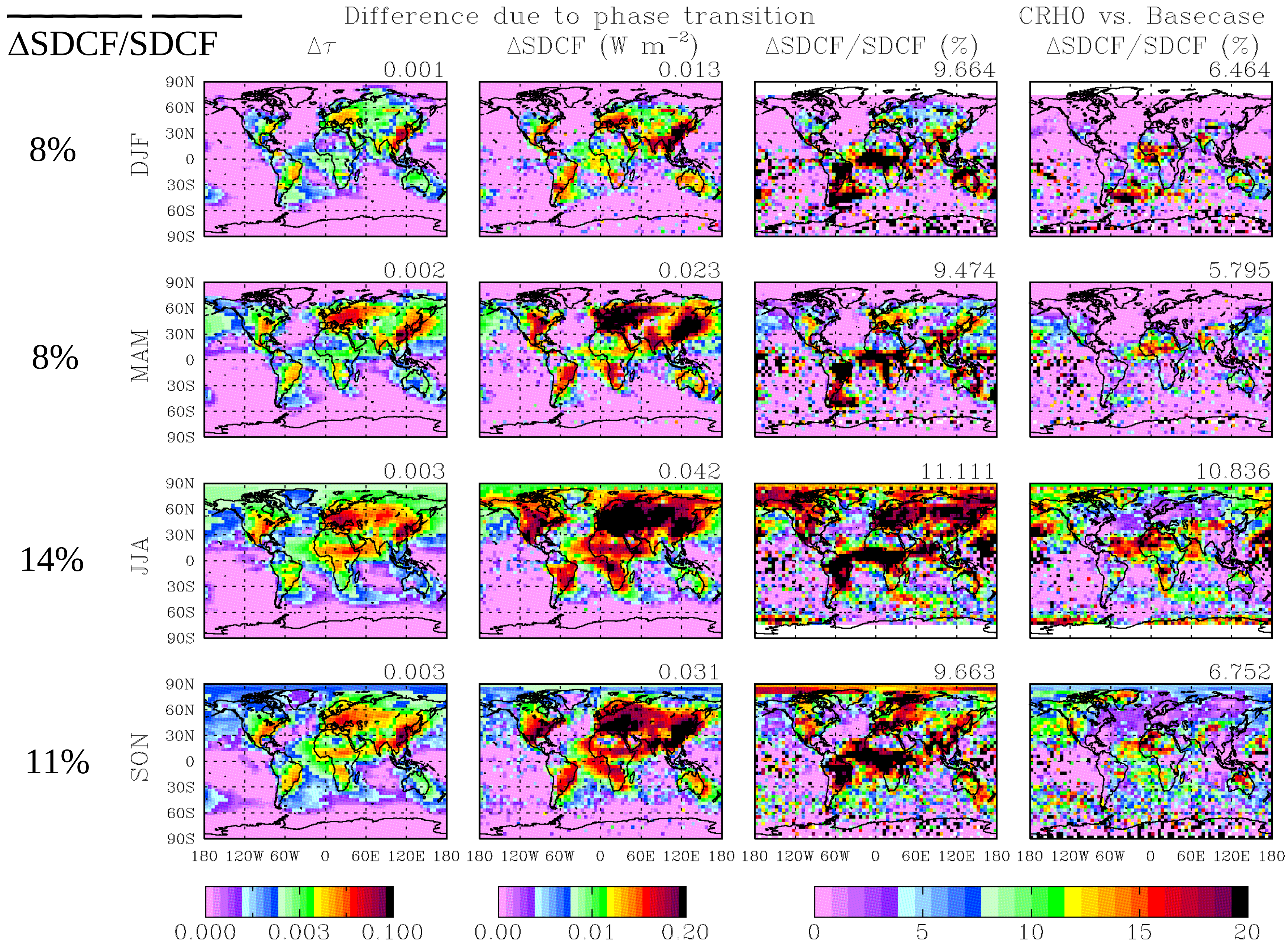
25%

18%



Forcing difference is over the Sahel region.

Effect of particle state on SDCF



Quantitative Summary of SDRE

	Basecase	CRH=0	CRH=DRH	DRH=CRH
B (mg SO4 m^{-2})				
Total	2.601	2.591	2.609	2.590
% due to solids	27.0%	0.0%	48.3%	12.7%
τ at 0.55µm (*10000)				
Total	240	253	217	247
% due to solids	15.4%	0.0%	30.0%	6.9%
$SDRE_{cld}$ (wm^{-2})				
Total	0.356	0.371	0.332	0.363
% due to solids	24.4%	0.0%	44.0%	11.8%
% to Basecase				
τ	100%	105.4%	90.4%	102.9%
$SDRE_{cld}$	100%	105.1%	93.0%	101.9%

Quantitative Summary of SDCF

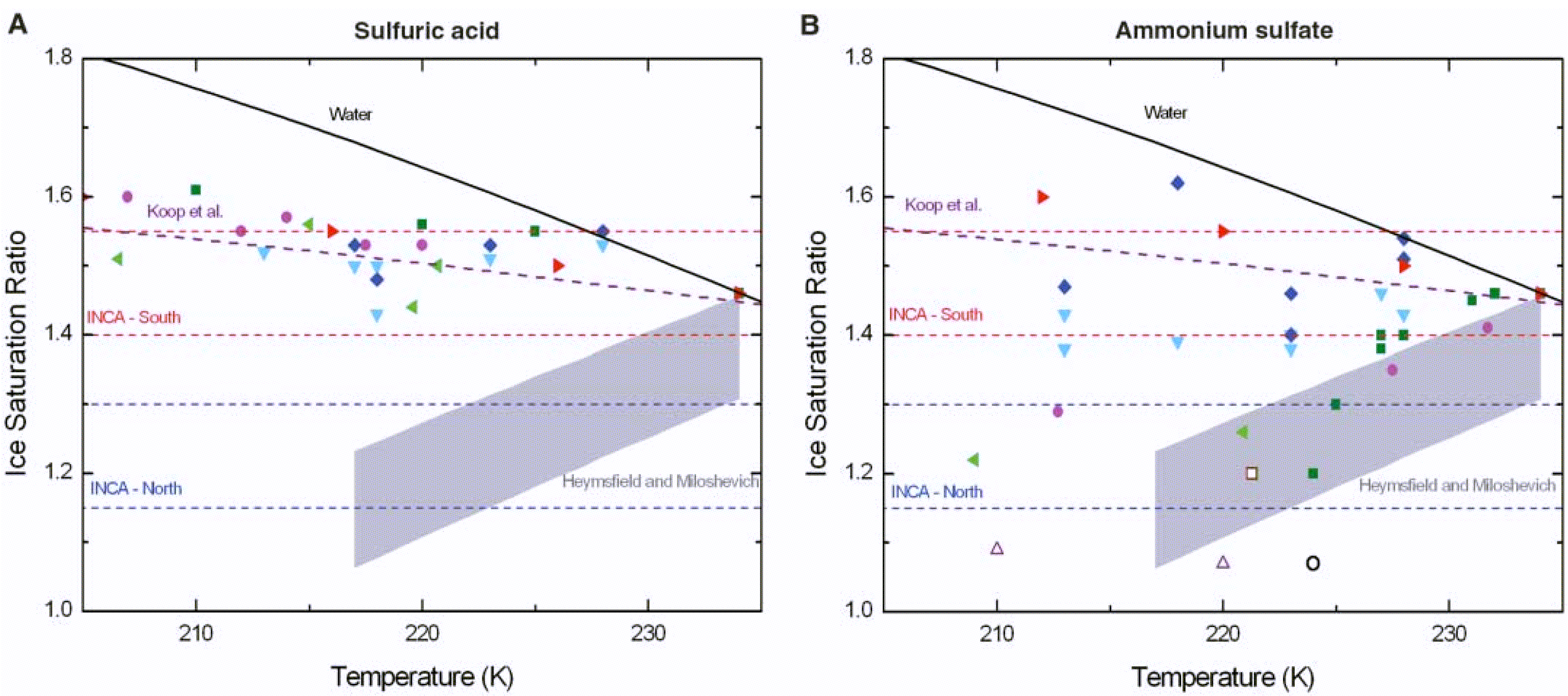
	basecase	CRH=0	CRH=DRH	DRH=CRH
B (mg SO4 m^{-2})				
Total	1.859	1.849	1.866	1.851
Solids %	28.1%	0%	52.4%	13.0%
τ at 0.55µm (*10000)				
Total	166	175	136	171
Solids %	16.9%	0%	36.8%	7.0%
SDCF (wm^{-2})				
Total	0.254	0.266	0.240	0.259
Solids %	25.6%	0%	47.5%	12.0%
Compared to base case				
τ % to basecase	100%	105.4%	81.9%	103.0%
$SDCF_{cld}$ %	100%	104.7%	94.5%	102.0%

Discussion

Solid Ammonium Sulfate Aerosols as Ice Nuclei: A Pathway for Cirrus Cloud Formation

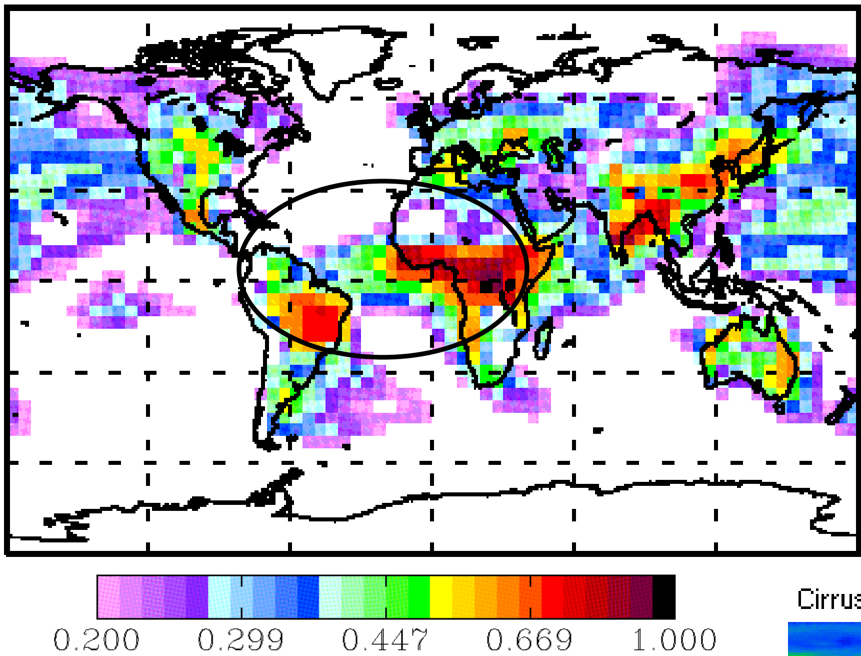
J. P. D. Abbatt,[1*] S. Benz,[2] D. J. Cziczo,[3] Z. Kanji,[1] U. Lohmann,[3] O. Möhler[2]

science, 2006



Sulfate phase impact on cirrus formation

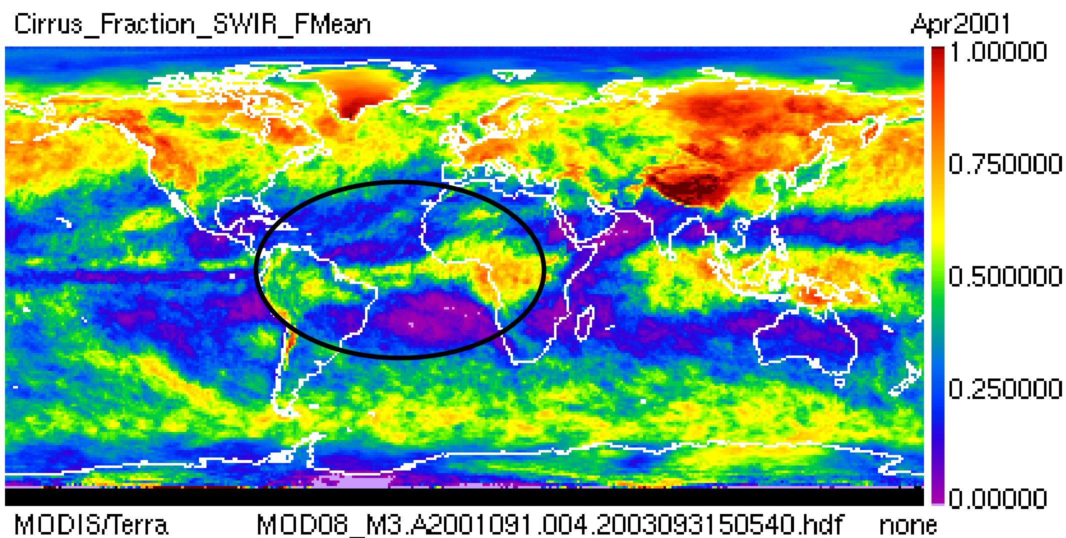
Regions where phase transition has the largest impact. $\Delta M_{sd}/M$



Places having important phase transition all show larger cirrus cloud fraction.

The reverse is not necessarily true, because there are other factors

Cirrus cloud fraction from MODIS



Summary

- Simulation of sulfate phase in H_2SO_4-H_2O system is developed. Inclusion of nitrate and organic aerosols is on the way.

- $SDCF_{CRH=0}$ is about 4% larger than $SDCF_{basecase}$, $SDCF_{CRH=DRH}$ is 6% smaller than $SDCF_{basecase}$.

- The percentage might vary if the model doesn't resolve the sulfate chemical composition, and hence hygroscopicity.

- Phase transition impact on SDCF has the important seasonal and regional variations, with larger effect (>20%) over south America, south Africa, east Asia, Europe and U.S. during summer time.

- The simulation results have important implications for understanding cirrus cloud formation.

Acknowledgement

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- NSF
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Assume same size distribution of dry particles, as RH changes from 0% to 80%, E increases by a factor of 2.7! For the same RH, chemical composition results in variation of E within 20%.

	RH=0				
	m_r	density (gcm^{-3})	r_{eff} (μm)	E m^2(g dry particle)$^{-1}$	E $m^2(gSO_4^{2-})^{-1}$
AS	1.53	1.76	0.17	3.85	5.31
AHS	1.47	1.78	0.17	3.29	3.95
LET	1.51	1.83	0.17	3.53	4.55
SA	1.84	1.84	0.17	3.42	3.48
Water	1.33	1.00			

3.52±0.24, ±7% 4.32±0.79, ±18%

	RH=80%				
AS	1.41	1.30	0.24	9.62	13.28
AHS	1.38	1.30	0.25	9.84	11.81
LET	1.40	1.32	0.25	9.85	12.71
SA	1.37	1.24	0.30	15.49	15.80

11.2±2.86, ±26% 13.4±1.7, ±13%

In GOES-CHEM, E ~ 11.5, assume rg=0.05, this study 0.07

Literature 8.0 - 16.0